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On the (MILD) combustion of gaseous, liquid, and solid fuels in high temperature preheated air

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Abstract

This paper examines the fundamental and industrial application aspects of combustion of natural gas, heavy and light fuel oils, and coal in highly preheated air. The experiments have been carried out in an experimental furnace at 0.58 MW thermal input based on fuel, and the combustion air has been preheated to 1300 °C. The fuel injectors are positioned outside of the combustion air stream. Detailed in-furnace measurements of temperature, chemistry (O₂, CO, NO_x, and particulates), and heat transfer have been performed. Combustion of natural gas and light oil takes place without a visible presence of flame. Although the furnace was operated with an overall excess air of 10%, the combustion process occurs in strongly sub-stoichiometric conditions due to entrainment of large amounts of recirculated flue gases into the fuel jets before ignition. The experiments demonstrated an effective technology for efficient and environmentally friendly combustion of a wide range of fuels. The technology discussed in the main text offers the potential of high furnace efficiencies, uniform heat flux distribution, and dramatic reductions in CO₂, CO, and NO emissions. Therefore, this technology should be considered for future design of industrial furnaces.

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Keywords: Mild combustion; Flameless combustion; High temperature air combustion

1. Introduction

In a recent publication from the authors, a new methodology for furnace design that offers the potential for substantial energy savings and dramatic reductions in CO₂, CO, and NO_x emissions was presented [1]. In this paper, further advances in the technology are discussed regarding the most important advances both in heat recovery methods and in NO_x reduction methodologies when combustion air with temperatures in

excess of 1000 °C are used. Regenerative burners that provide preheated combustion air at the prerequisite high temperature are essential elements of the technology [2–4]. The NO_x reduction is achieved by positioning the fuel injectors at an optimum radial distance from the combustion airstream, a method that has been pioneered by Tokyo Gas [5] and advocated since 1990s by the International Flame Research Foundation (IFRF) [1,6]. The essential features of this new combustion technology can be summarized as follows:

- (a) combustion air is preheated to temperatures in excess of 1000 °C and in some applications up to 1300 °C,

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- (b) the furnace exit temperature (before the regenerator) is only slightly higher (50–100 °C) than the combustion air temperature,
- (c) the fuel nozzles are positioned at an optimum radial distance from the combustion air-stream injecting the fuel into hot recirculated combustion products with low oxygen concentration, typically 2–5 volume %,
- (d) both the fuel jets and the combustion air jet entrain large quantities of combustion products before their mixing takes place,
- (e) combustion takes place globally throughout the furnace, and typically no flame is visible. The in-furnace temperatures are relatively uniform with only small gradients appearing in close proximity to the burner. The same is applicable to the in-furnace oxygen field,
- (f) the radiative fluxes are uniform.

2. The technology

There is no doubt that Japan was the leading country to implement this energy-efficient and environment-friendly combustion technology. Originally, the technology was named Excess Enthalpy Combustion [7] while today it is called High Temperature Air Combustion (HTAC) in Japan, Flameless Oxidation (FLOX) in Germany, Low NO_x Injection (LNI) in the USA, and Mild Combustion (MILD) in Italy.

A review on the development of the various high temperature air combustion processes has been provided by Weber et al. [1], while Katsuki and Hasegawa [4] gave a comprehensive review of the fundamental aspects related to the development of the technology (see also Tsuji et al. [8]).

3. Some fundamentals of high temperature air combustion

In previous work by Tanaka et al. [3] and Gupta et al. [9], it was observed that a substantial increase in radiation intensity at wavelengths corresponding to C₂ and CH radical emissions with preheated air under low oxygen concentration conditions took place. Japanese companies have already developed a flame visualization technique based on the luminescence intensity ratio of C₂ to CH radicals. In 1997, the IFRF carried out semi-industrial-scale experiments [1,10] that identified principal characteristics of the Excess Enthalpy Combustion of natural gas. The Laser Sheet Visualization Technique, using Mie scattering, was combined with Laser Doppler Anemometry and intrusive (probes) for temperature and chemistry (CO, CO₂, O₂, and NO) measurement.

These techniques allowed for a comprehensive mapping of the in-furnace combustion process. High levels of entrainment of flue gas were observed for both air and natural gas jets. The measured radiative heat fluxes at the furnace walls in this work were high and uniform. The uniformity and homogenization has also been recently observed by Ishiguro et al. [11].

Plessing et al. [12] used laser-induced pre-dissociative fluorescence and Rayleigh thermometry to examine flameless oxidation at laboratory scale. De Joannon et al. [13] have examined the applicability of the existing chemical reaction schemes for combustion of hydrocarbons to high temperature air combustion conditions. In their most recent publication, they argue that flameless oxidation can be described as a two-stage combustion in which the first part is in rich conditions with a significant excess of inert gases.

Little is known about oil combustion with high temperature air. Shimo [14] studied emission spectra of liquified petroleum gas (LPG) and kerosene flames in a laboratory-scale combustor. Carbon dioxide, nitrogen, and argon have been used to dilute the high temperature air (around 1000 °C) and by doing so to simulate combustion products of low oxygen content. While bright luminous flames have been observed when ambient air is used, green flames have been seen at 5% oxygen concentration. The green colouration has been associated with enhanced C₂-radical emissions. In reducing thermal NO_x emissions, carbon dioxide has been demonstrated to be more effective than nitrogen. The authors have attributed this behaviour to the higher specific heat capacity of carbon dioxide. In 1998, the IFRF had carried out semi-industrial scale tests using both a light and a heavy fuel oil [15,16]. Realization of flameless combustion for the light oil was straightforward. However, in the case of heavy fuel oil the results were not so clear, and particulate emissions of around 400 mg/Nm³ were observed, and the flames were luminous.

There have been only two significant attempts to apply this technology to combustion of pulverized coal. Kiga et al. [17] carried out laboratory-scale tests in a drop tube-furnace burning a high volatile coal with high-temperature, low oxygen-content air. Their measurements indicated that increasing air preheat resulted in increased combustion efficiency and reduced NO_x emissions. It was concluded in this work that “the use of high-temperature, diluted air was not suitable for pulverized coal combustion.” The results of the study presented here somewhat contradict these conclusions. In 1999, the IFRF conducted semi-industrial scale experiments that revealed a high potential for this new technology for the combustion of solid fuels as well as gaseous and liquid fuels [18].

4. Objectives of the present work

The IFRF has carried out a series of semi-industrial scale tests using highly preheated air. Natural gas [1,10], light and heavy fuel oils [15,16], and a single coal [18] were used as fuels. These experiments were carried out under almost identical fuel input conditions with the same air preheat, identical firing density, and similar heat extraction rates. The objectives of the current work were to examine the potential of this new technology for efficient and environment-friendly combustion of various fuels from an experimental viewpoint.

5. Experimental

The experiments were executed in a refractory lined furnace at The IFRF, see Fig. 1 of [19]. The furnace has a 2 m × 2 m cross-section, and its length is 6.25 m. The furnace was equipped with one burner operated under steady state conditions. The high temperature regenerator was replaced by a precombustor where natural gas was burned with air under fuel lean conditions. Oxygen was added to the hot gases exiting the precombustor to maintain a concentration of 21%. Such a comburent (vitiated air) simulates the air preheat of the excess enthalpy combustion pro-

cess. The experimental burner operates at 0.58 MW fuel input and high air preheat see, Table 1. The vitiated air is supplied through the central channel with an injection velocity of 85 m/s. As can be seen from the tables, the typical NO levels entering the system with the preheated air before the main combustion process occurs are of the order of 100 ppm vd as measured.

The experimental burner was operated so that the fuel is supplied using the two horizontal injectors/atomizers. Thus, the overall flow pattern, temperature, and chemistry fields are 3-dimensional. The in-furnace measurements, reported in this paper, have been obtained by traversing the furnace in the horizontal plane of the two fuel injectors.

6. Results and analysis

6.1. Visual observations

While firing natural gas or a light fuel oil with highly preheated (vitiated) air, it is difficult to visually determine where the combustion takes place since the whole furnace glows without any visible presence of flames. In contrast with the natural gas and light oil flames, the heavy fuel oil flames and pulverized coal flames were clearly distinguishable from the wall radiation.

Table 1
Experimental conditions

	Flow rate (kg/h)	Temp (°C)	Enthalpy (MW)	Composition % (vol)
<i>Natural gas (NG)</i>				
Natural gas	47	25	0.58	CH ₄ 87.8%, C ₂ H ₆ 4.6%, C ₃ H ₈ 1.6%, C ₄ H ₁₀ 0.5%, LCV = 44.76 MJ/kg
Vitiated air (comburent)	830	1300	0.35	19.5% wet O ₂ , 59.1% wet N ₂ , 15% H ₂ O, 6.4% wet CO ₂ , 110 ppm vd NO
Furnace exit gases	877	1220	0.380	1.6% wet O ₂ , 140 ppm vd NO
<i>Light fuel oil (LFO)</i>				
Light oil	49	25	0.58	C 86.56%, H 13.43%, S 0.074%, N < 0.05%, LCV = 42.66 MJ/kg
Vitiated air (comburent)	800	1270	0.33	19.6% wet O ₂ , 60.1% wet N ₂ , 14% H ₂ O, 6.4% wet CO ₂ , –70 ppm vd NO
Furnace exit gases	849	1130	0.325	1.1% wet O ₂ , 95 ppm vd NO
<i>Heavy fuel oil (HFO)</i>				
Heavy oil	51	105	0.58	C 87.98 %, H 10.48 %, S 1.08%, N 0.37%, LCV = 40.75 MJ/kg
Vitiated air (comburent)	765	1320	0.33	20.1% wet O ₂ , 59.7% wet N ₂ , 14% H ₂ O, 6.6% wet CO ₂ , 70 ppm vd NO
Furnace exit gases	816	1240	0.350	1.1% wet O ₂ , 215 ppm vd NO
<i>Coal</i>				
High volatile Guasare coal	66	—	0.58	C 78.41%, H 5.22%, O 10.9 S 0.82%, N 1.49%, Ash 3.3%, Volatiles 37.1%, Fixed carbon 57.7%, Moisture 2.9%, LCV = 31.74 MJ/kg
Vitiated air (comburent)	675	1350	0.30	19.7% wet O ₂ , 57.2% wet N ₂ , 15.1% H ₂ O, 8.1% wet CO ₂ , 110 ppm vd NO
Furnace exit gases	871	1230	0.360	2.6% wet O ₂ , 220 ppm vd NO

6.2. In-furnace measurements

6.2.1. Flow field

The flow field in the furnace is determined by the strong, central jet of the combustion airstream. This large momentum jet drives a strong reverse flow zone that recirculates hot combustion products back towards the furnace front wall region. The two fuel jets are directly injected into the recirculation zone. The fuel jets entrain a substantial amount of hot gases of rather low-oxygen content (1–3%) before mixing with the combustion airstream. For example, the natural gas jet increases its original mass of 23.5 kg/h into 225 kg/h within a distance of 0.6 m (around 50 nozzle diameters) downstream of the injection location. The oil jets entrain only half of the natural gas jet entrainment. The central combustion airstream triples its original mass within a distance of 0.3 m (around 25 nozzle diameters). The implication of this observation is that the combustion of gaseous fuels (natural gas, evaporated oils, and volatile matter) as well as individual oil droplets and coal particles takes place in a low-oxygen content environment.

6.2.2. Temperature

Figure 1 shows the measured in-furnace temperatures. The first observation is apparent; although the combustion air temperature is higher than 1270 °C, the peak temperatures within the furnace do not exceed 1550 °C. Second, the temperature gradients are much lower than in “conventional” firing. Only in the region local to the fuel jets do steep gradients appear, and therefore one observes a rather homogeneous temperature field. The whole furnace is filled with combustion products with temperatures in the range of 1300–1500 °C. The furnace exit temperatures are 1220, 1130, 1240, and 1230 °C for natural gas, light fuel oil, heavy fuel oil, and coal firing, respectively. The peak temperature is reached at the boundary between the central and the fuel jets, where intense combustion takes place. It is important to notice that the fuel jets emerge into the recirculating combustion products at a temperature circa 1250–1300 °C, and the fuel is oxidized in a low oxygen environment.

6.2.3. Chemistry

Detailed in-furnace measurements of oxygen concentration reveal that, with the exception of the near burner region, gradients in primary gas composition profiles are relatively smooth and flat. It was observed that the whole furnace volume is filled up with combustion products containing 2–3% oxygen. Carbon monoxide has been detected almost three-quarters of the distance downstream from the burner into the furnace for all the four fuels fired. The LFO-flame

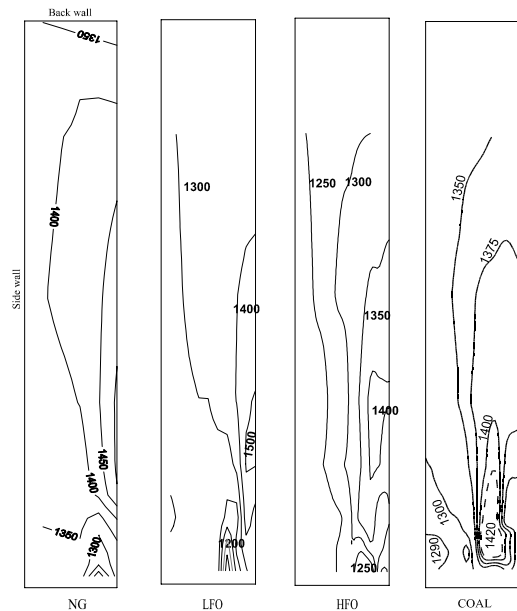


Fig. 1. In-flame temperature distributions (temperatures in °C).

shows substantially larger CO concentration than the HFO-flame. For the LFO-flame CO concentrations up to 2% were found at an axial distance of 2 m. The HFO-flame shows a the highest CO concentration of 1.7% at an axial distance of 80 cm, and at 2 m the CO concentration had dropped to values below 0.4%. It is somewhat surprising that the CO concentrations in the LFO case are so much higher than in the HFO case. While firing coal, CO concentrations up to 5% have been measured in the fuel jet. No traces of CO have been found in the chimney. For NG-, LFO-, and HFO-flames, hydrogen is found in the first half of the furnace, and its presence indicates the slow combustion as well as the cracking of the fuel. For the NG-flame, peak values of 2.5% H₂ concentrations have been found, while figures of 3% and 6% are applicable for LFO- and HFO-combustion, respectively.

6.2.4. Pollutants

Whilst considering the NO emissions, one should remember that a substantial amount of NO_x is being brought into the furnace with the combustion airstream. Figure 2 shows the generalized flue gas NO_x emissions for all fuels fired.

The NO “contamination” level of the combustion airstream is as high as 110 ppm for the NG-case while figures of 70 and 110 ppm are applicable to the LFO-, HFO-, and coal-cases, Fig. 2. For the natural gas firing and the light oil firing, the net increase in the NO emissions is 30 and 5 ppm, respectively. The experiments show a lower furnace exit NO emission as well

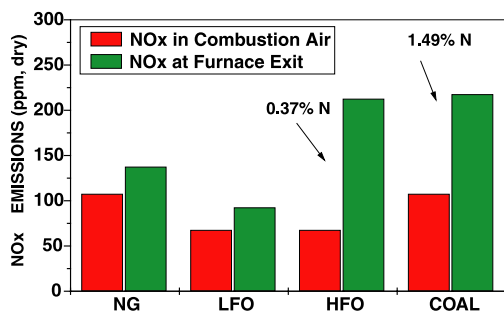


Fig. 2. Generalized NO_x emissions. For each fuel, the left bar is for NO_x in combustion air and the right bar for NO_x at furnace exit.

as a lower net NO production when firing a light oil rather than natural gas. We attribute this difference to the fact that the LFO experiments were carried out under slightly different thermal conditions than the NG experiments. In the LFO case, the furnace exit temperatures were around 90 °C lower than the case for natural gas, see Table 1. Notwithstanding the possibility of reducing some of the inlet NO (entering the furnace with the combustion airstream) by the NO_x reburning mechanisms, it is most likely that the thermal NO_x mechanism is the dominant one. Figure 3 shows the in-furnace measured NO_x concentrations (typically NO constitutes 95% of the measured NO_x the remainder being NO₂). In both the NG case and the LFO case

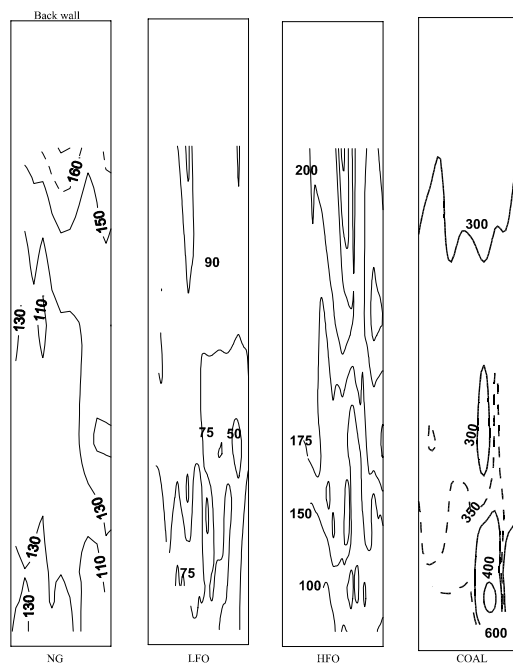


Fig. 3. In-flame NO_x measurements.

are the results are somewhat similar; no regions of high NO_x formation rates can be identified, and most of the NO formation seems to occur in the downstream part of the furnace.

Figures 2 and 3 show very interesting results when both HFO and coal are combusted and the results compared (the nitrogen content of the fuels are 0.37% and 1.49%, respectively). The furnace exit NO_x emissions are in the range 215–220 ppm (at 3% O₂) for both fuels. While firing coal, it was possible to reduce the emissions further to 150–170 ppm by optimizing the fuel jet positions and the momentum. Conventional firing of these fuels in the IFRF Furnace No.1 would normally result in NO_x emissions of the order of 400–600 ppm (HFO) and 800–1000 ppm (coal) due to the fuel NO formation mechanism. It would be possible to obtain a 60–70% NO_x emission reduction by applying standard low NO_x burner technology consisting of either air or fuel staging techniques. Figure 2 shows that much lower NO_x emissions are achievable by application of the combustion technology demonstrated in this work. It is important to realize that the furnace NO emissions of 215–220 ppm have been obtained without any considerations given to reducing fuel NO. The contours of the measured NO_x concentrations, Fig. 3, indicate that a reburning mechanism in which NO is reduced to molecular nitrogen is effective in the case of coal. One may argue that the presence of the same mechanism in the HFO case is also evident. Simplistically, one may reason further on the fact that the low NO_x emissions with coal firing are due to the NO reburning mechanism prevailing under high temperatures and a relatively long residence time (around 10 s). However, this is certainly an over-simplified explanation. It is rather well known that the fuel NO formation mechanism is not so sensitive to temperatures, and therefore it has been in general, accepted that recirculating combustion products (flue gases) are not effective in fuel NO reduction. However, in the light of the present HFO and coal results, one should not underestimate the role the recirculating flue gas plays in generating locally sub-stoichiometric conditions favourable for the NO_x reburning mechanism.

A discussion on NO_x emissions should be carried out in parallel with considerations on carbon monoxide emissions, soot and particulate emissions and char burnout (carbon in ash). While firing NG, no CO emissions were observed when the oxygen content at the furnace exit was maintained above 1%. When a HFO was fired [15,16] particulate emissions in excess of 400 mg/Nm³ were observed. In a previous publication [16], we identified the need for designing oil atomizers suitable for the process. The most recent trials confirmed that a family of new atomizers performed extremely well, reducing

the particulate emissions to a level of typically 15–20 mg/Nm³. A high volatile coal (Guasare) was used in our trials, and a high burnout in excess of 99.7% was observed.

6.2.5. Total radiative heat flux

The NG and HFO measurements were performed after allowing the furnace to stabilize for a week, resulting in a flue gas temperature of about 1250 °C for both the NG and HFO flames. The flue gas temperature of the LFO flame was lower (~1130 °C) since the furnace did not reach a steady state at the time of the measurements. The heat flux profiles are flat for all the fuels. The heat flux values vary from about 310 kW/m² for the LFO to about 375 W/m² for the HFO and coal. The NG values are in between at around 330 kW/m². Had the furnace reached full steady state while firing the light oil, it is considered that the heat fluxes for light oil and natural gas combustion would be similar.

7. Conclusions

In this work, combustion of natural gas (NG), light fuel oil (LFO), heavy fuel oil (HFO), and coal with 1300 °C comburent containing 20% oxygen, 60% nitrogen, 14% water vapour, and 6% carbon dioxide was examined. The combustion process of light oil was very similar to that of natural gas. The entire furnace was visible illuminated, with no evidence of visibly flames being observed. However, combustion of heavy fuel oil and coal was significantly different, and the flames were always visible. The following conclusions are applicable to this work:

- (a) Due to a slow combustion process, the temperature and the chemistry fields are uniform throughout the experimental furnace. CO was not found in the furnace exit for any of the experimental fuels.
- (b) Axially, along the whole furnace, a high radiative heat flux of 300–400 kW/m² was measured. The flatness of the measured heat flux indicates that the combustion takes place throughout the furnace.
- (c) Although the furnace has been operated with the overall excess air of 10%, the combustion of evaporated oils and coal volatile matter occurs in strongly sub-stoichiometric conditions.
- (d) Very interesting and significant results were obtained when a high volatile coal (1.49% N) was combusted with high temperature air. Also in this case, low combustion rates were observed under locally sub-stoichiometric conditions. The lowest NO_x emissions were

in the range 160–175 ppm (at 3% O₂), indicating the high NO reduction potential of the technology for nitrogen containing fuels.

Acknowledgments

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Comments

Antonio Cavaliere, University of Naples, Italy. The relatively poorer performances of burner with heavy oil fuels could be related to lower momentum of normal injection. Have you some assessments about velocities of the different fuels?

Reply. The injection velocity of the natural gas at the furnace front wall is 60 m/s. The injection velocity for coal (transport air velocity) has been varied in the range from 26 to 43 m/s. The coal injectors have been positioned 175, 280, and 385 mm away from the burner center. For obtaining minimum NO_x emissions an optimization of both the injection velocity and the injector position is needed. The injection average velocity of the heavy fuel oil droplets was measured to be 60 m/s and the D_{10} diameter was 35 μm . The measurements were performed using a Phase Doppler Particle Analyzer. The oil injector was positioned 280 mm away from the burner center and nitrogen was used as atomizing

medium to decrease the oxygen availability. We believe that under the above specified conditions the oil spray jets entrain substantially less flue gas than the natural gas jet.

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Alan N. Sayre, Babcock and Wilcox, USA. In suspension firing systems for power generation, the membrane wall heat absorption is much higher than your experimental system and the combustion air temperatures are about 600 K. Can this technology be extended to such systems by the addition of oxygen into the primary air to stabilize the pulverized coal flame?

Reply. Application of this technology to suspension firing for power generation is not straightforward. The main difficulty is in obtaining such a high level of air pre-heat. Your suggestion is very attractive, indeed.